Thin Film Li-La-Zr-O Electrolyte for Li Metal Cells

Joong Sun Park[1], Lei Cheng[1], Jordi Cabana[1], Guoying Cheng[1], Marca Doeff[1], Thomas J. Richardson[1], Wan-Shick Hong[2].

1. Lawrence Berkeley National Laboratory, Berkeley, California, USA; 2. University of Seoul, Seoul, Republic of Korea.

Batteries based on lithium metal as a negative electrode could potentially bring about significant increases in energy density over current lithium-ion technologies. However, several concerns including stability, safety, and cycling efficiency have limited their development. The tendency of lithium metal to form mossy deposits or dendrites upon cell recharge leads to unsafe cells and short lifetimes. The use of solid state protective layers, which conduct ions but are electronically insulating, has been proposed as a potential solution to improve cycle life and enable its use with high energy air and sulfur cathodes.[1] Several phases such as LISICON and $La_{0.5}Li_{0.5}TiO_3$, among others, are being investigated for this purpose.[2, 3] However, these compounds are not stable with respect to reduction by lithium and may form electronically conducting phases. To prevent this, an interlayer between the lithium electrode and solid ionic conductor is required. Phases that do not contain redox active transition metals are preferable to avoid the need for such an interlayer. We have recently studied the lithium ion-conducting phosphosilicate phase, $Li_{3.4}Si_{0.4}P_{0.6}O_4$, which meets this criterion. $Li_{3.4}Si_{0.4}P_{0.6}O_4$ forms a stable interface against lithium and has an ionic conductivity of 4.5×10^{-6} S/cm at room temperature.4 Similarly, garnet $Li_7La_3Zr_2O_{12}$ (LLZO) ceramic electrolyte appears to be stable against lithium metal electrodes, and has been reported to have high ionic conductivity (3×10^{-4} S/cm).[5] Approaches for preparing thin, pinhole-free, dense layers are needed to ensure good current capability. The goal is to minimize the ceramic electrolyte impedance while preventing contact between the lithium metal anode and a liquid electrolyte, which wets the cathode.

To this end, we are investigating the fabrication of $Li_7La_3Zr_2O_{12}$ thin films using pulsed laser deposition (PLD). The films were characterization by employing X-ray diffraction, laser induced breakdown spectroscopy (LIBS), rutherfold backscattering spectroscopy (RBS), and electrochemical impedance spectroscopy. Here we will discuss the physical and electrochemical properties of $Li_7La_3Zr_2O_{12}$ films fabricated with different crystalline phase. Chemical stability of the film against lithium metal will be discussed by employing X-ray photoelectron spectroscopy (XPS).

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References

[1] M-Y Chu, S. Visco, L.C. De Jonghe, U.S. Patent No. 6402795, 2004.

[2] H.S. Zhou, Y.G. Wang, Chem Commun 46 (2010) (34) 6305.

[3] Y. Inaguma, C. Liquan, M. Itoh, T. Nakamura, T. Uchida, H. Ikuta, M. Wakihara, Solid State Commun. 86 (1993) 689.

[4] L. Zhang, L. Cheng, J. Cabana, G. Chen, M. M. Doeff, and T. J. Richardson, Solid State Ionics 231 (2013) 109.

[5] R. Murugan, V. Thangadurai, and W. Weppner, Angew. Chem. Int. Ed., 46 (2007) 7778